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INVESTIGATION OF THE ABSORPTION OF INFRARED RADIATION BY NITROUS OXIDE FROM 4000 to 6700 cm^{-1} (2.5 to 1.5 μm)

by

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INTRODUCTION AND SUMMARY

As part of a large program to tabulate the parameters of all the significant absorption lines of atmospheric gases, we have recently analyzed some N_2O data in the 4000-6700 cm^{-1} region obtained a few years ago on a different project. The bands in this region are generally much weaker than the fundamental bands and many of the combination bands that occur at lower wavenumbers. It seems unlikely that a band with strength less than 10^{-21} $molecules^{-1}cm^{2}cm^{-1}$ would absorb significantly over any usable atmospheric path; therefore, we have restricted the careful analysis to the bands above this "cut-off" value. Approximate strengths and upper limits have also been determined for several of the weaker bands, but further study would be required to determine their strengths accurately.

Douglas and Moller[1] and Plyler, Tidwell and Allen[2] have identified several of the N_2O bands in the region of interest and have published data on the line positions. These workers did not include information on the strengths of lines or bands. Pliva[3,4] has accumulated previous data on N_2O energy levels and has tabulated many of the energy levels and constants from which line positions can be determined accurately. In a previous report[5], we listed the strengths of a few of the bands contained in this study; however, the previous report did not include the detailed curves and tables shown below.

From data on the absorption on the high wavenumber side of the head of the $00^{o}3$ band, we have found that the extreme wings of N_2O lines are quite sub-Lorentzian; i.e., they absorb less than Lorentz-shaped lines with the same strengths and widths.

EXPERIMENTAL PROCEDURES AND DATA ANALYSIS

The spectral curves were scanned with a grating spectrometer and strip-chart recorder with the spectral resolution varying from approximately 0.2 to 0.9 cm^{-1}. Several of the curves were digitized, and a computer was used to calculate the transmittance T, absorptance A, $(-1/u) \ln T$, and $(-1/u)\int \ln T \, d\nu$. Samples were contained in a multiple-pass absorption cell with path lengths, L, up to 3290 cm. Since the cell had been cooled to 196 K for a different project, a few transmittance curves were obtained for samples at this temperature. These curves are valuable in identifying the temperature-sensitive difference bands which result from transitions from an excited vibrational level. Samples at pressures less than 1 atm were employed to study the line structure within the bands. In order to obtain information about band strengths, we used $N_2O + N_2$ samples at approximately 15 atm so that the structure was smoothed out. Under this condition, the observed transmittance is very nearly equal to the actual transmittance that would be observed with infinite resolving power. The quantity $(-1/u)\ln T$ is then equal to the absorption coefficient, and the integral of this quantity over a spectral interval is the sum of the strengths of the bands within the interval. The absorber thickness u is expressed in molecules of N_2O/cm^2. The quantity $(-1/u)\ln T$ is essentially independent of pressure for a wide range of pressures greater than about 10 atm, which is required to smooth out the line structure. Data from references 1, 2, and 4 were used to identify the absorption bands and to

determine line positions. Pliva's article is more recent than the other two and incorporates the results of several previous articles. Therefore, Pliva's values for energy levels were used in preference to others when they were available.

SPECTRAL DATA AND BAND STRENGTHS

Figures 1-15 show spectral curves for the regions containing the stronger bands between 4000 and 6700 cm^{-1}. Two curves are shown for most of the regions. The first is a curve of absorptance for samples with line structure, and the second is of $(-1/u) \ln T$ for higher-pressure samples. Band identifications and comments on the curves appear in Table 1.

The three fundamental bands ν_1, ν_2, and ν_3 are at 1284.907, 588.767, and 2223.756 cm^{-1}, respectively. Note that $\nu_3 \cong 2\nu_1 \cong 4\nu_2$. The quantity N defined as $2v_1 + v_2 + 4\ v_3$, where the v's are the vibrational quantum numbers, is convenient in specifying energy levels and in estimating band positions. Because of the approximate relationship between ν_1, ν_2, and ν_3, levels having the same N are approximately equal. It follows that bands arising from transitions from the 00^00 level to different levels having the same N occur near each other. All of the important N_2O bands between 4000 and 6700 cm^{-1} resulting from transitions from the 00^00 state have upper levels with N between 7 and 12.

Table 1 lists all of the bands with the 00^00 lower level and upper levels from N = 7 to N = 12. Values of the band centers followed by P and PTA are from Pliva[4] and Plyler, Tidwell and Allen[2], respectively. The centers of several of the bands with N of 10, 11, or 12, are not given by either of these authors and have not been calculated since they are too weak to be

of interest in atmospheric transmission problems. We note that for N = 7, 8, and 9, the band at the lowest wavenumber in each group has the largest v_3 and smallest v_1. As expected, the highest wavenumber band has the smallest v_3 and largest v_1. Also included in Table 1 are the numbers of the figures in which spectral curves of the bands can be seen. The remarks column contains additional information about the observance of the bands. All of the stronger bands ($S_v > 1$ E-21 molecules^{-1}cm^{2}cm^{-1}) can be seen in the figures. A few very weak bands were observed in spectral regions between those covered by the figures. The raw data for these very weak bands were not analyzed.

The strengths of several of the bands were determined by integrating $(-1/u)\ln T$ over the spectral interval including the band. As discussed previously, the curves of $(-1/u)\ln T$ were based on samples at high enough pressure for the structure to be smoothed out. The interval integrated over for a particular band also contains associated difference bands arising from transitions from excited vibrational states with the same changes in vibrational quantum numbers as the band of primary interest. The difference bands are weaker because of the lower population of the excited states. Strengths listed in Table 1 include the difference bands associated with the fundamental or combination band. In a few cases, it was necessary to account for overlapping by other bands. Care was exercised in measuring the bands stronger than 10^{-21} molecules^{-1}cm^{2}cm^{-1}. With the exception of the 23^10 band at 4335.798 cm^{-1}, these stronger bands were measured with reasonable accuracy. More spectral data with good resolution are required in order to account for the overlapping of this band with neighboring ones. Upper limits and approximate strengths were determined for some of the weaker bands. Those indicated with an approximate sign (~) may be in error by as much as a factor of 2 or 3.

Additional information about the relative strengths of lines within the bands and of the different branches can be obtained from Table 2 which lists the cumulative integral of $(-1/u)\ln T$. Values are tabulated each

5 cm^{-1} and near the centers of most of the strong bands. The value of the integral between any two wavenumbers listed can be determined by subtracting the corresponding values of the cumulative integral. Spectral regions containing only very weak bands are not included in the table.

LINE SHAPE

A few years ago, we[6] investigated the absorption on the high-wavenumber side of the head of the 00^o3 band of CO_2 near 7000 cm^{-1}. From the absorption data and previous knowledge of the strengths and widths of the lines, we were able to infer the shapes of the extreme wings of the lines centered on the low wavenumber side of the band head. We found that self-broadened CO_2 lines absorbed less beyond about 5 cm^{-1} from the line centers than Lorentz-shaped lines with the same widths and strengths. Lines broadened by N_2 deviated even further from the Lorentz shape.

A similar study has been made near 6600 cm^{-1} on the high wavenumber side of the head of the 00^o3 N_2O band shown in Figs. 14 and 15. Between 6600 and 6650 cm^{-1}, most of the absorption by samples at pressures greater than a few atm is due to the extreme wings of the lines centered between 6500 and 6600 cm^{-1}. The shapes of the N_2O lines inferred from these data are surprisingly similar to those found earlier for CO_2 lines. The difference between the results from self broadening is less than the experimental uncertainty. The same is also true for N_2-broadened lines.

TABLE 1

BAND IDENTIFICATIONS AND STRENGTHS

Band	Band Center (cm^{-1})	Fig. No.	Strength ($molecules^{-1} cm^2 cm^{-1}$)	Remarks
				N = 7
$0\ 7^1\ 0$	4037.13 P		<5 E-23	Not apparent in Fig. 1. Possibly masked by 11^11 band.
$1\ 5^1\ 0$	4197.960 P		<5 E-23	Not observed.
$2\ 3^1\ 0$	4335.798 P	3	~1 E-21	Observed in raw data. Some of R branch in Fig. 3.
$3\ 1^1\ 0$	4446.379 P		<1 E-22	Q branch observed in raw data.
$0\ 3^1\ 1$	3931.258 P		~1 E-22	Observed in raw data.
$1\ 1^1\ 1$	4061.979 P	1	1.10 E-21 ±5%	Difference bands also appear in Fig. 1.
				N = 8
$0\ 8^0\ 0$	4601.80 P		<1 E-21	Possibly masked by 12^01 band.
$1\ 6^0\ 0$	4767.13 P		<2 E-21	Not observed.
$2\ 4^0\ 0$	4911.06 P	8	6.5 E-22 ±10%	
$3\ 2^0\ 0$	5026.34 P	9	2.9 E-21 ±12%	Overlaps 01^12 band.
$4\ 0^0\ 0$	5105.65 P	10	2.9 E-21 ±10%	
$0\ 4^0\ 1$	4491.541 P		~2 E-22	Observed in raw data.
$1\ 2^0\ 1$	4630.164 P	5	6.8 E-21 ±10%	
$2\ 0^0\ 1$	4730.828 P	6	4.4 E-20 ±10%	
$0\ 0^0\ 2$	4417.379 P	3	6.9 E-20 ±10%	Difference band also appears in Fig. 3.
				N = 9
$0\ 9^1\ 0$	5168.27 P	11		Q branch may show in Fig. 11.
$1\ 7^1\ 0$	5338.51 P		<2 E-22	Not observed.
$2\ 5^1\ 0$	5489.74 P		<2 E-22	Not observed.
$3\ 3^1\ 0$	5617.85 P		<2 E-22	Not observed.
$4\ 1^1\ 0$	5722.90 P		<2 E-22	Not observed.

TABLE 1 (Continued)

Band	Band Center (cm^{-1})	Fig. No.	Strength ($molecules^{-1} cm^2 cm^{-1}$)	Remarks
				N = 9 (Contd.)
$0\ 5^1\ 1$	5053.582 P		<1 E-22	Possibly masked by 32^00 and 40^00 bands.
$1\ 3^1\ 1$	5200.780 P		$\lesssim$2 E-22	Not observed.
$2\ 1^1\ 1$	5319.175 P		$\lesssim$2 E-22	Not observed.
$0\ 1^1\ 2$	4977.695 P	9	~5 E-22	Q branch is prominent. Overlaps 32^00 band.
				N = 10
$0\ 10^0\ 0$			$\lesssim$2 E-22	Not observed.
$1\ 8^0\ 0$			$\lesssim$2 E-22	Not observed.
$2\ 6^0\ 0$			$\lesssim$2 E-22	Not observed.
$3\ 4^0\ 0$			$\lesssim$2 E-22	Not observed.
$4\ 2^0\ 0$	6295.06 PTA		~3 E-22	Observed in raw data.
$5\ 0^0\ 0$			$\lesssim$2 E-22	Not observed.
$0\ 6^0\ 1$			$\lesssim$2 E-22	Not observed.
$1\ 4^0\ 1$			$\lesssim$2 E-22	Not observed.
$2\ 2^0\ 1$	5887.99 PTA		~4 E-22	Observed in raw data.
$3\ 0^0\ 1$	5974.74 PTA	13	1 E-21	
$0\ 2^0\ 2$				Not observed.
$1\ 0^0\ 2$	5646.59 PTA	12	1 E-21	
				N = 11
$0\ 11^1\ 0$				None of the bands for N = 11 were observed. The band centers are expected to occur between 6200 and 7000 cm^{-1}. Their strengths are $\lesssim$2 E-22.
$1\ 9^1\ 0$				
$2\ 7^1\ 0$				
$3\ 5^1\ 0$				
$4\ 3^1\ 0$				
$5\ 1^1\ 0$				

TABLE 1 (Continued)

Band	Band Center (cm^{-1})	Fig. No.	Strength ($molecules^{-1} cm^2 cm^{-1}$)	Remarks
				N = 11 (Contd.)
0 7^1 1				
1 5^1 1				
2 3^1 1				
3 1^1 1				
0 3^1 2				
1 1^1 2				
				N = 12
0 12^0 0				The 00^03 band is the only band for N = 12 that was observed. Most of the others are probably centered above 6700 cm^{-1}, the upper limit of the region studied.
1 10^0 0				
2 8^0 0				
3 6^0 0				
4 4^0 0				
5 2^0 0				
6 0^0 0				
0 8^0 1				
1 6^0 1				
2 4^0 1				
3 2^0 1				
4 0^0 1				
0 4^0 2				
1 2^0 2				
2 0^0 2				
0 0^0 3	6580.83 PTA	14	1.52 E-21 ± 6%	

TABLE 2

$$-\frac{1}{u}\int_{\nu'}^{\nu} \ln T \, d\nu$$

(Molecules^{-1} cm^{2} cm^{-1})

(Multiply all Values by 10^{-24})

ν' = 3990

ν (cm^{-1})	
4000	2.30
4005	5.91
4010	11.24
4015	19.39
4020	30.97
4025	48.89
4030	73.90
4035	111.7
4040	162.5
4045	228.8
4050	297.2
4055	405.5
4060	562.5
4062^c	706.0
4065	837.7
4070	904.9
4075	975.5
4080	1041.1
4085	1084.3
4090	1102.9
4095	1107.3
4100	1107.5

ν' = 4290

ν (cm^{-1})	
4300	13.7
4305	31.8
4310	56.7
4315	89.2
4320	128.5
4325	170.1
4330	223.4
4335	319.9
4340	466.3
4345	664.7
4350	918.4
4355	1245.2
4360	1696.3
4365	2352.7
4370	3333.6
4375	4722.8
4380	6586.1
4385	8841.4
4390	11682.
4395	15699.
4400	21285.
4405	27782.
4410	33828.
4415	37638.
4417^c	38390.
4420	39582.
4425	44559.
4430	53452.
4435	62958.
4440	68757.
4445	69734.
4450	69793.
4455	69822.
4460	69842.

ν' = 4560

ν (cm^{-1})	
4565	5.0
4570	16.9
4575	40.0
4580	79.4
4585	146.2
4590	250.9
4595	410.3
4600	650.3
4605	987.3
4610	1420.6
4615	1939.9
4620	2549.0
4625	3130.6
4630^c	3506.4
4635	3854.7
4640	4507.0
4645	5335.4
4650	6061.3
4655	6494.9
4660	6686.7
4665	6789.4

ν' = 4665

ν (cm^{-1})	
4670	105.8
4675	243.8
4680	443.7
4685	772.9
4690	1319.0
4695	2235.7
4700	3617.8
4705	5614.3
4710	8367.7
4715	11823.
4720	15644.
4725	19044.
4730	21032.
4731^c	21271.
4735	22758.
4740	27380.
4745	34334.
4750	40705.
4755	43614.
4760	43927.
4765	43965.
4770	43988.

ν' = 4850

ν (cm^{-1})	
4860	1.9
4865	5.5
4870	12.4
4875	25.0
4880	45.3
4885	76.1
4890	118.3
4895	170.5
4900	223.6
4905	269.8
4910	302.9
4911^c	307.9
4915	334.5
4920	391.8
4925	464.4
4930	532.7
4935	584.7
4940	622.9
4945	647.7
4950	676.6
4955	695.5
4960	717.9
4965	745.3
4970	782.5
4975	867.5
4980	1005.3
4985	1057.9
4990	1140.1
4995	1255.7
5000	1407.0
5005	1599.9
5010	1843.0
5015	2119.6
5020	2376.1
5025	2535.4
5026^c	2553.6

ν' = 4850

ν (cm^{-1})	
5030	2642.4
5035	2891.4
5040	3285.2
5045	3704.7
5050	4004.4
5055	4142.1
5060	4187.8
5065	4219.5
5070	4268.6
5075	4347.1
5080	4466.8
5085	4635.3
5090	4853.4
5095	5098.8
5100	5323.9
5105	5464.0
5106^c	5480.4
5110	5572.5
5115	5846.5
5120	6263.0
5125	6655.2
5130	6886.3
5135	6953.6
5140	6973.6
5145	7005.3
5150	7044.8
5155	7076.9
5160	7093.7
5165	7099.0
5170	7101.9
5175	7104.7
5180	7106.8

ν' = 5565

ν (cm^{-1})	
5570	1.3
5575	3.2
5580	5.4
5585	8.4
5590	13.4
5595	22.1
5600	35.8
5605	54.8
5610	80.8
5615	117.8
5620	116.0
5625	225.8
5630	300.8
5635	389.7
5640	477.4
5645	535.1
5647^c	544.6
5650	571.8
5655	668.9
5660	829.3
5665	976.6
5670	1006.
5675	1007.

ν' = 5916.5

ν (cm^{-1})	
5920	4.5
5925	13.0
5930	22.8
5935	39.1
5940	64.9
5945	101.9
5950	153.7
5955	221.8
5960	302.8
5965	387.9
5970	456.5
5975^c	496.9
5980	558.7
5985	685.0
5990	862.1
5995	999.4
6000	1035.
6005	1044.
6010	1045.

ν' = 6500

ν (cm^{-1})	
6505	5.2
6510	14.7
6515	26.7
6520	39.4
6525	56.2
6530	77.0
6535	98.6
6540	124.6
6545	168.7
6550	243.3
6555	344.2
6560	431.2
6565	534.4
6570	647.5
6575	745.9
6580	801.0
6581^c	808.3
6585	860.0
6590	1031.
6595	1328.
6600	1520.
6605	1520.

^{c} This indicates that ν is near the center of the primary band.

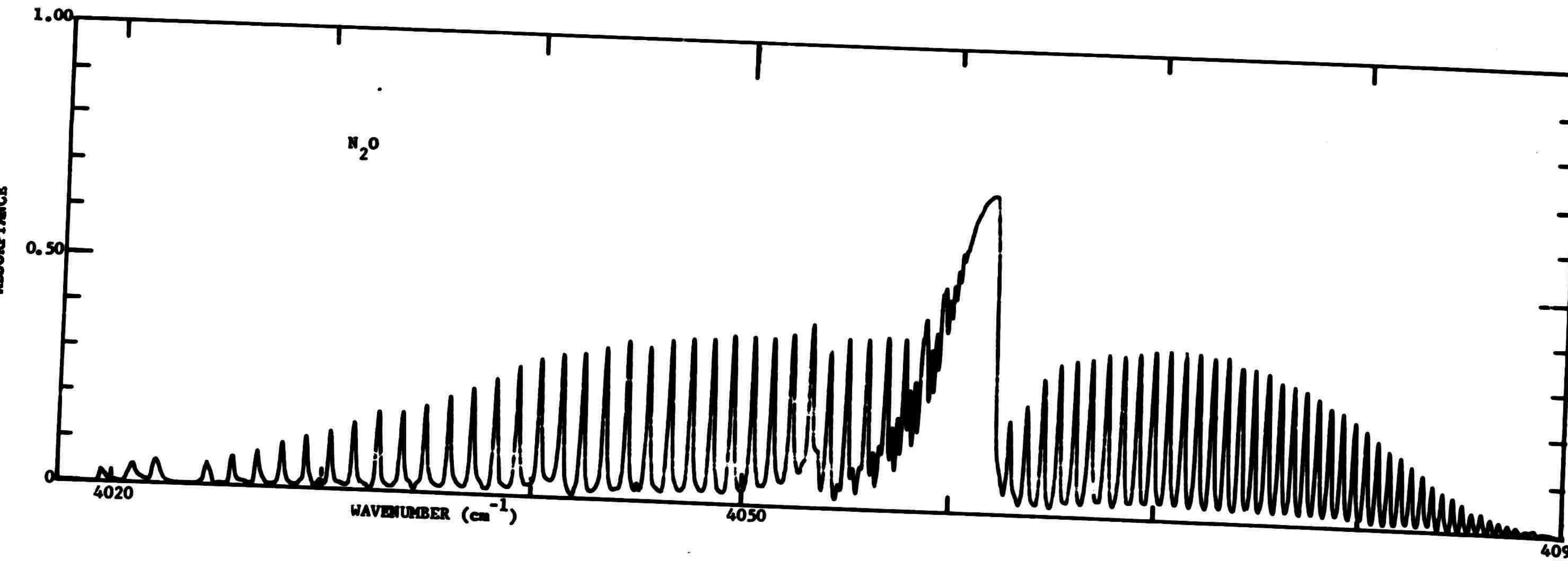

FIG. 1. Spectral curve of absorptance between 4020 and 4090 cm^{-1} for a pure N_2O sample. u = 163E20 molecules cm^{-2}; p = 0.132 atm; L = 3290 cm; θ = 196 K. Spectral slitwidth $\cong$ 0.25 cm^{-1}.

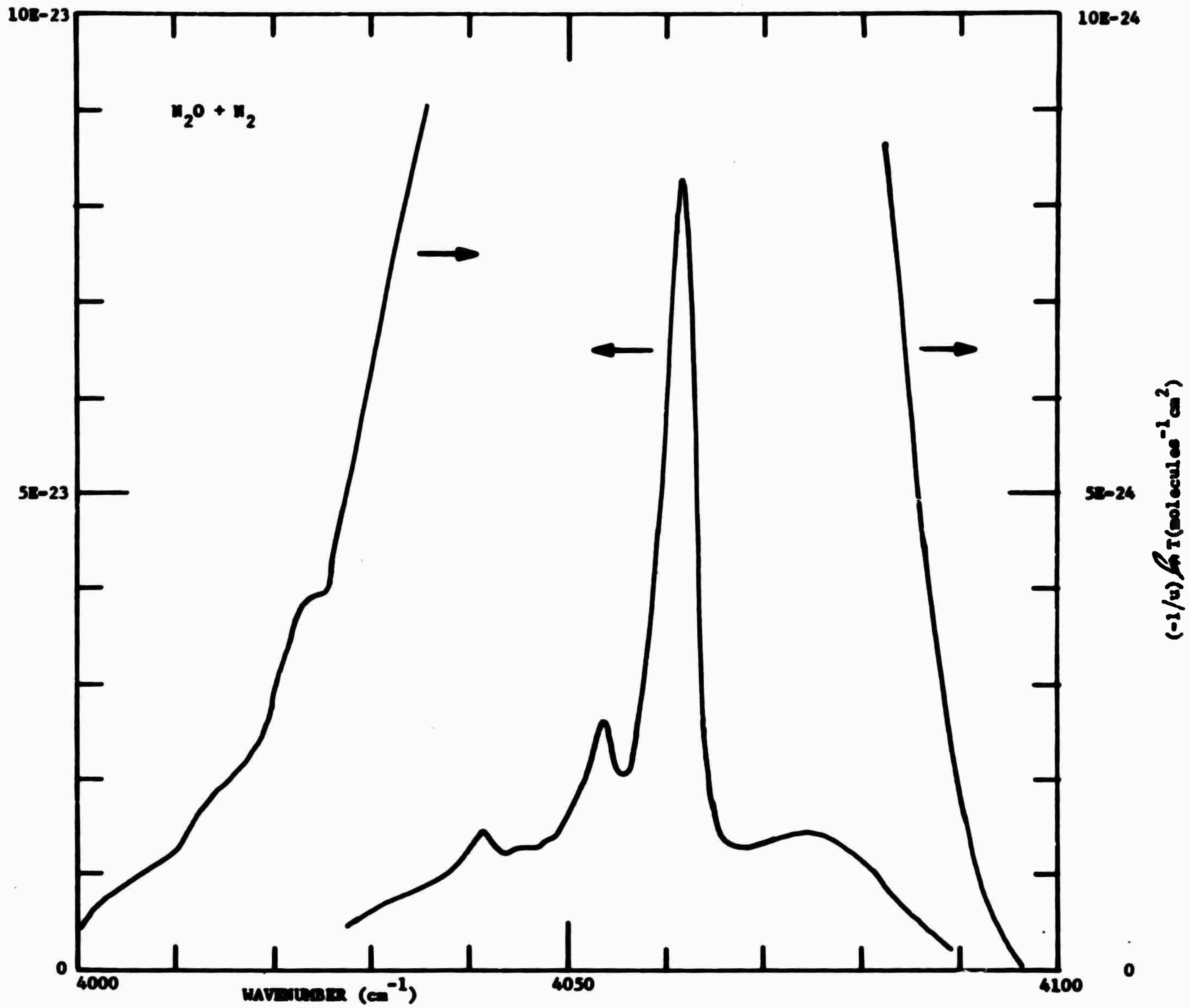

FIG. 2. Spectral curve of $(-1/u) \ln T$ between 4000 and 4100 cm^{-1} for an $N_2O + N_2$ sample at approximately 15 atm. θ = 296 K. Spectral slitwidth $\cong 0.25$ cm^{-1}. The arrows indicate the ordinate scale to be used.

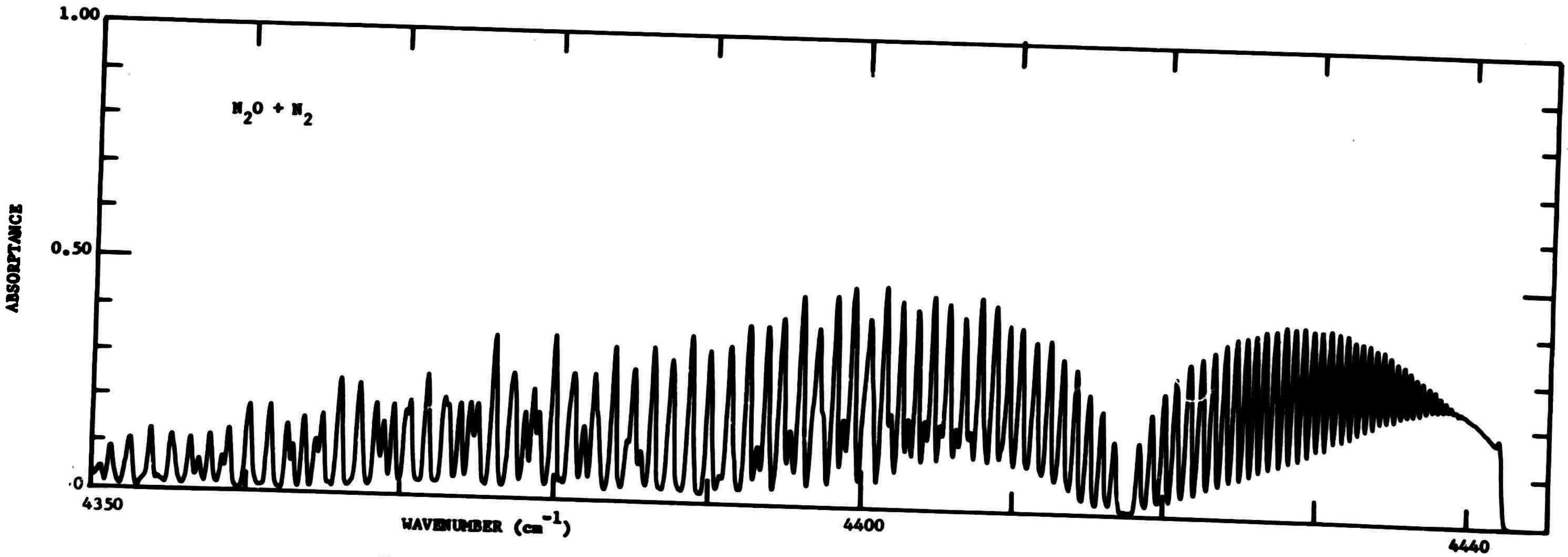

FIG. 3. Spectral curve of absorptance between 4350 and 4445 cm^{-1} for an $N_2O + N_2$ sample. u = 8.08 E20 molecules cm^{-2}; p = 0.0395 atm; P = 0.0947 atm; L = 826 cm; θ = 296 K. Spectral slitwidth $\cong$ 0.33 cm^{-1}.

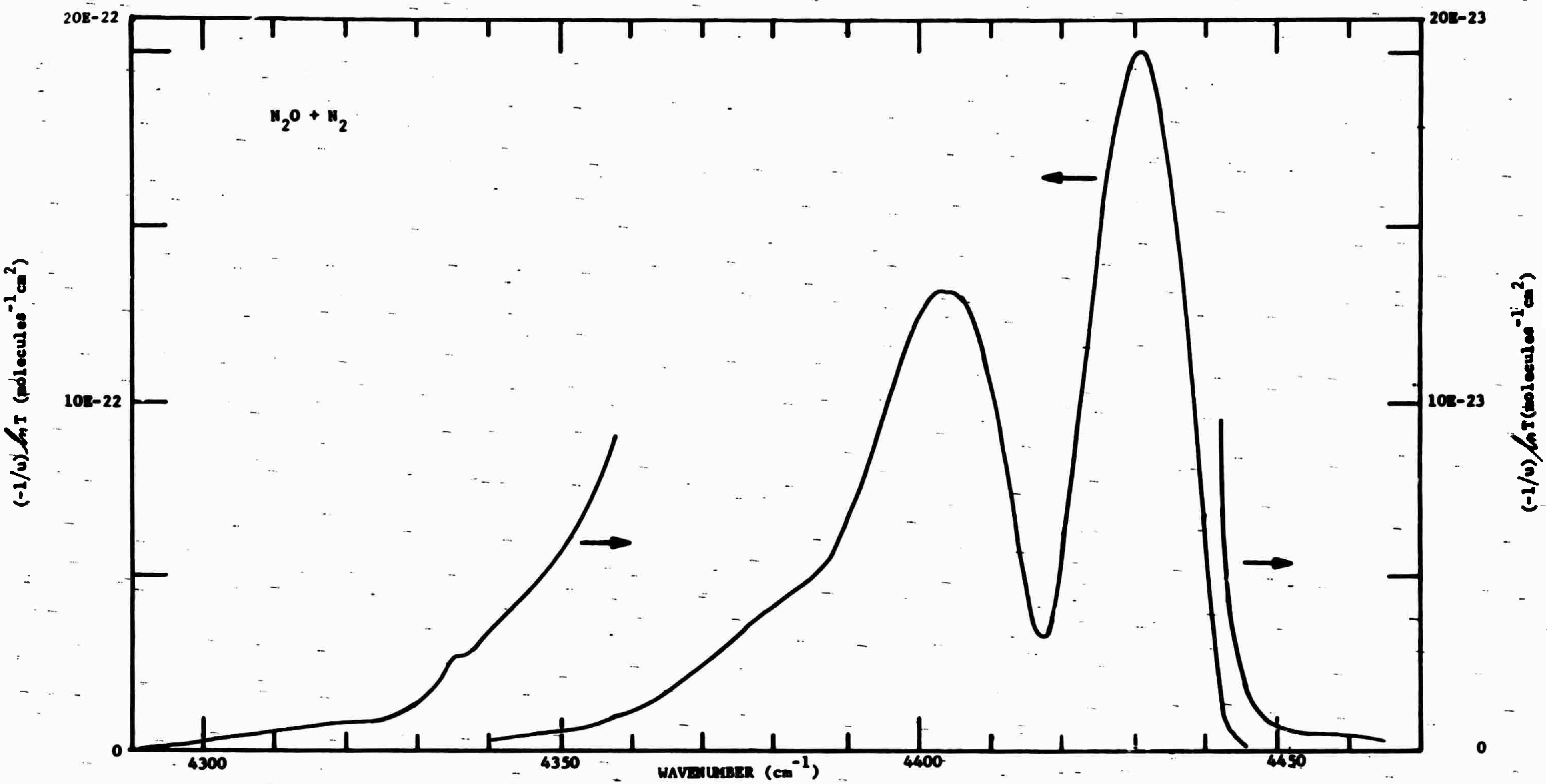

FIG. 4. Spectral curve of $(-1/u)\ln T$ between 4290 and 4465 cm^{-1} for an $N_2O + N_2$ sample at approximately 15 atm. θ = 296 K. Spectral slitwidth ≅ 0.33 cm^{-1}. The arrows indicate which ordinate scale to be used.

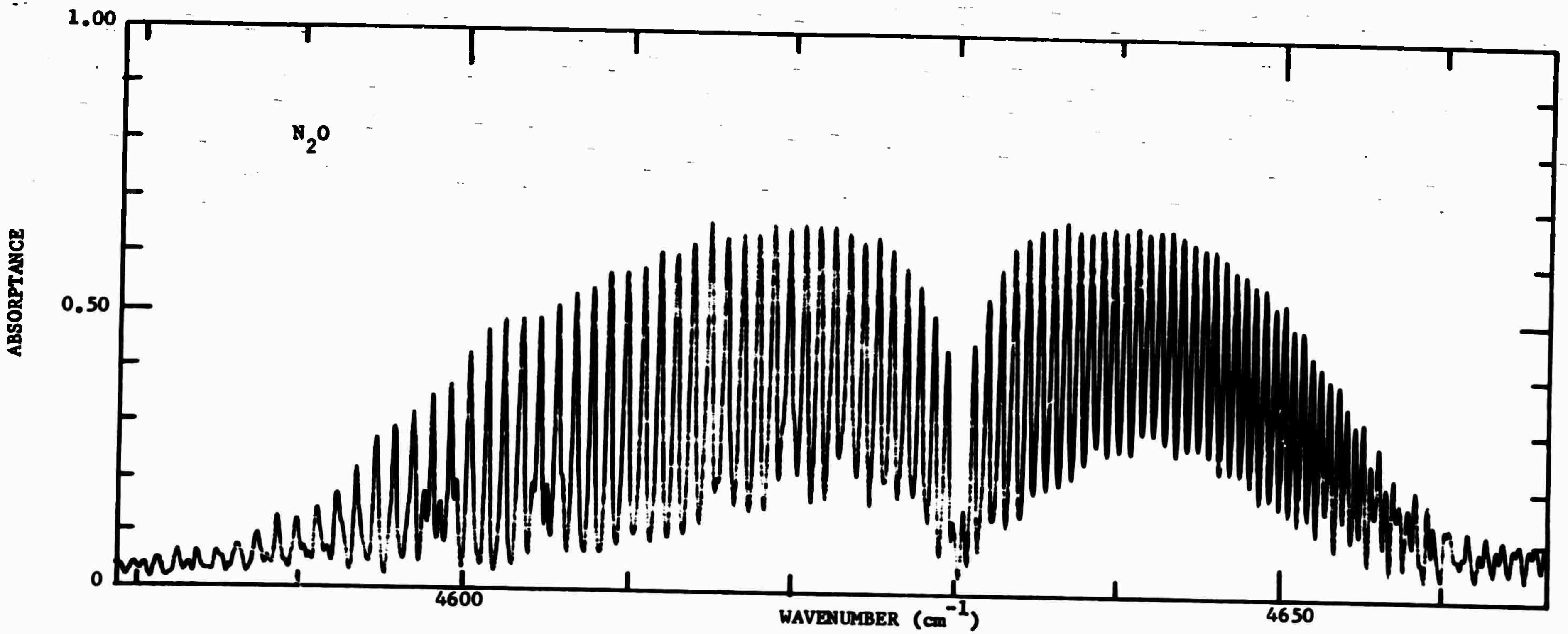

FIG. 5. Spectral curve of absorptance between 4580 and 4665 cm^{-1} for a pure N_2O sample. u = 163E20 molecules cm^{-2}; $P_1 = 0.132$ atm; $L = 3290$ cm; $\theta = 196$ K. Spectral slitwidth $\simeq 0.40$ cm^{-1}.

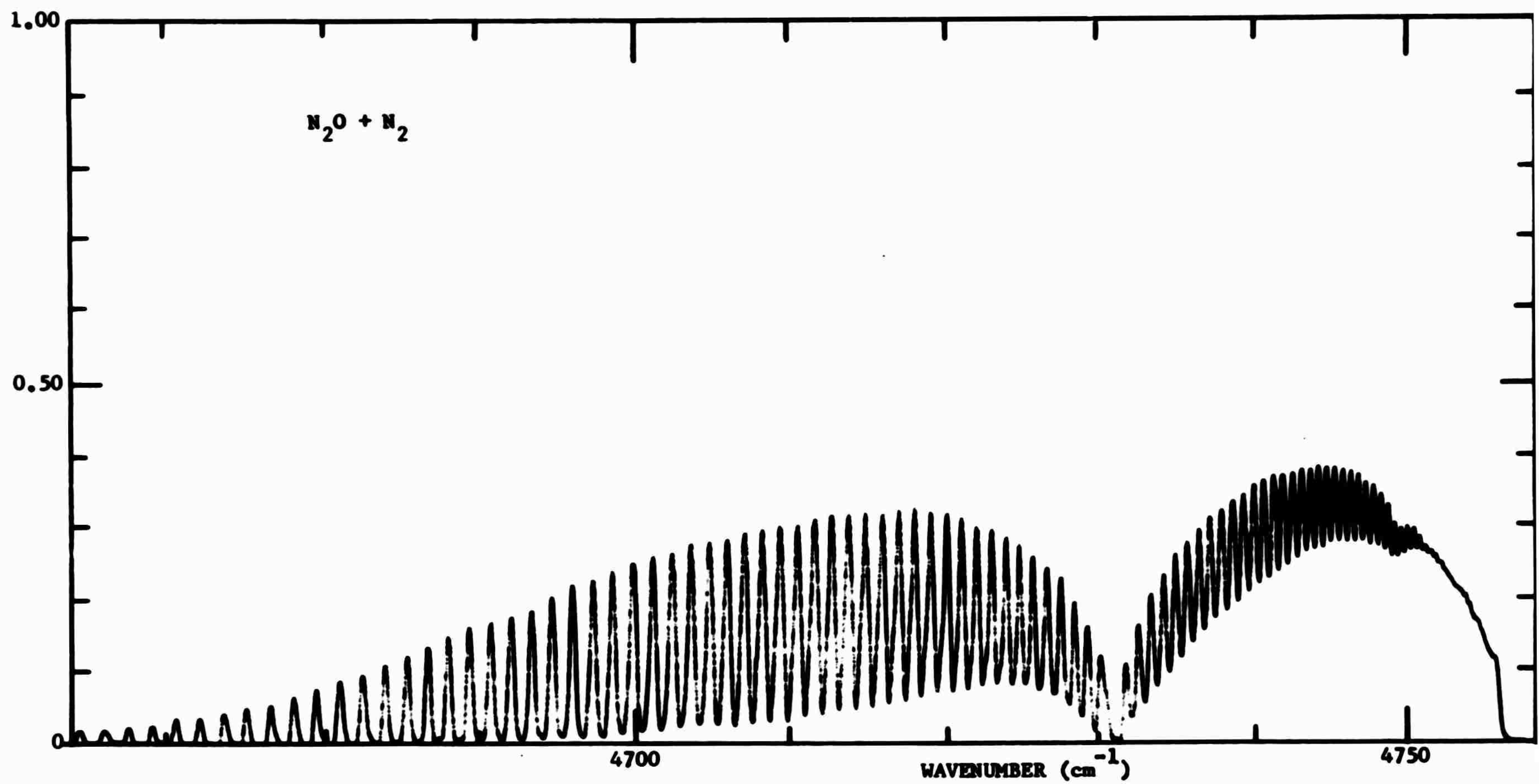

FIG. 6. Spectral curve of absorptance between 4665 and 4760 cm^{-1} for an $N_2O + N_2$ sample. $u = 8.08E20$ molecules cm^{-2}; $p = 0.0395$ atm; $P = 0.0947$ atm; $L \cong 826$ cm; $\theta = 296$ K. Spectral slitwidth $\cong 0.40$ cm^{-1}.

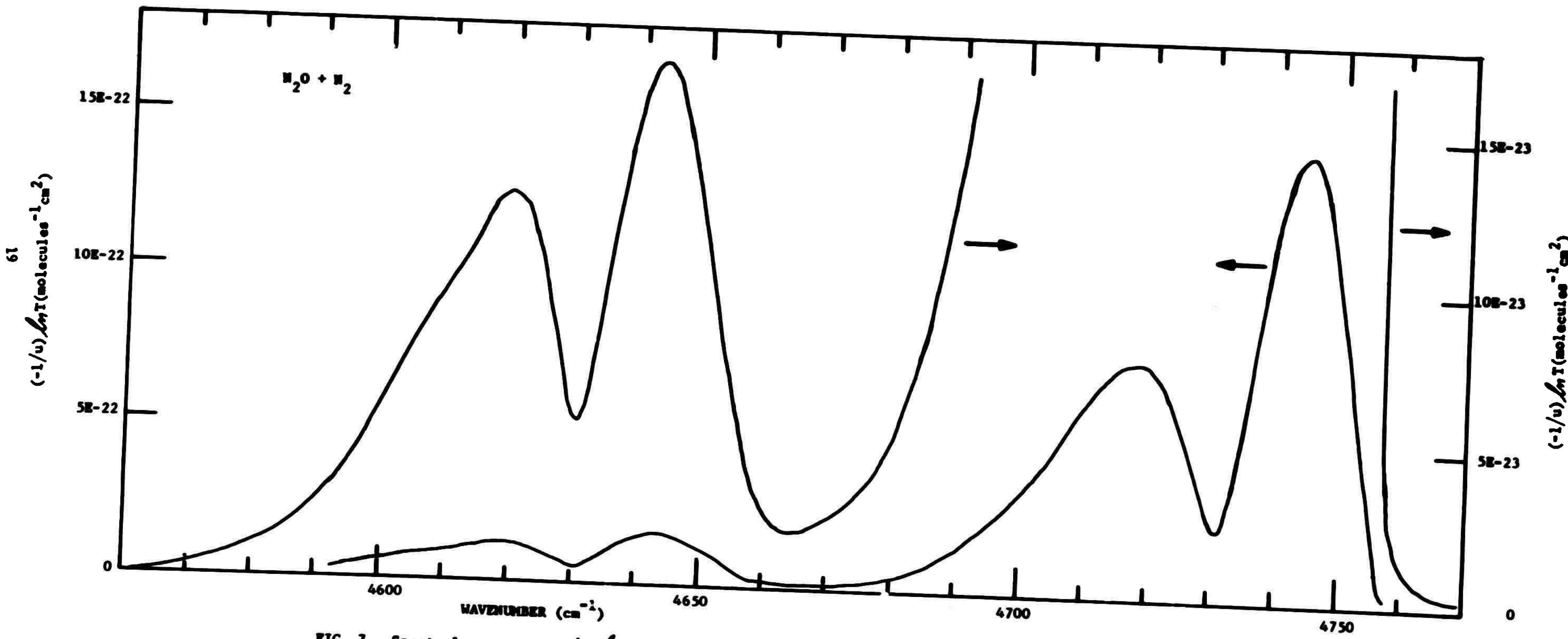

FIG. 7. Spectral curve of $(-1/u) \ln T$ between 4560 and 4770 cm^{-1} for an $N_2O + N_2$ sample at approximately 15 atm. θ = 296 K. Spectral slitwidth $\cong$ 0.38 cm^{-1}. The arrows indicate the ordinate scale to be used.

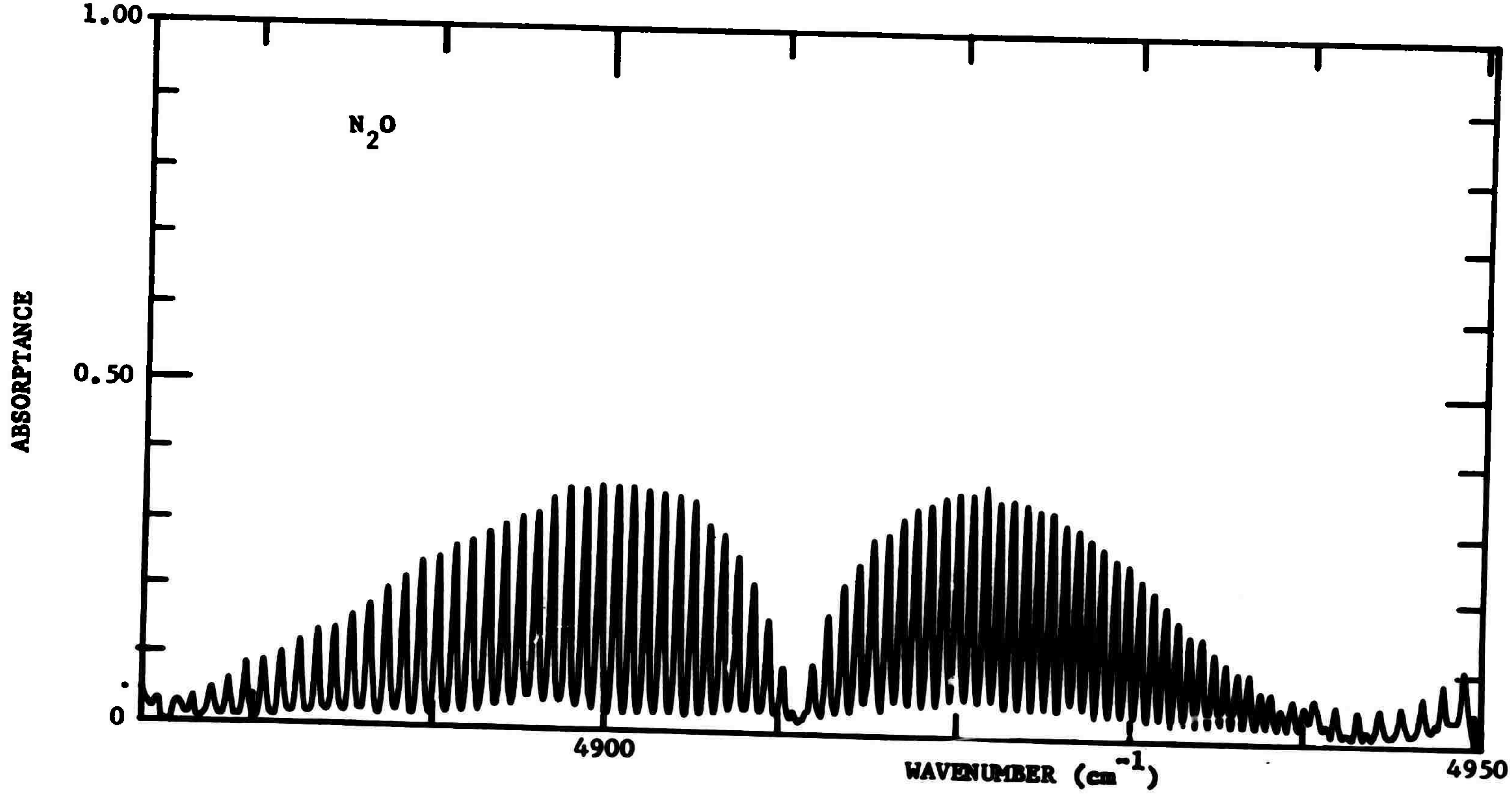

FIG. 8. Spectral curve of absorptance between 4875 and 4950 cm^{-1} for a pure N_2O sample. u = 163E20 molecules cm^{-2}; p = 0.132 atm; L = 3290 cm; θ = 196 K. Spectral slitwidth $\cong$ 0.35 cm^{-1}.

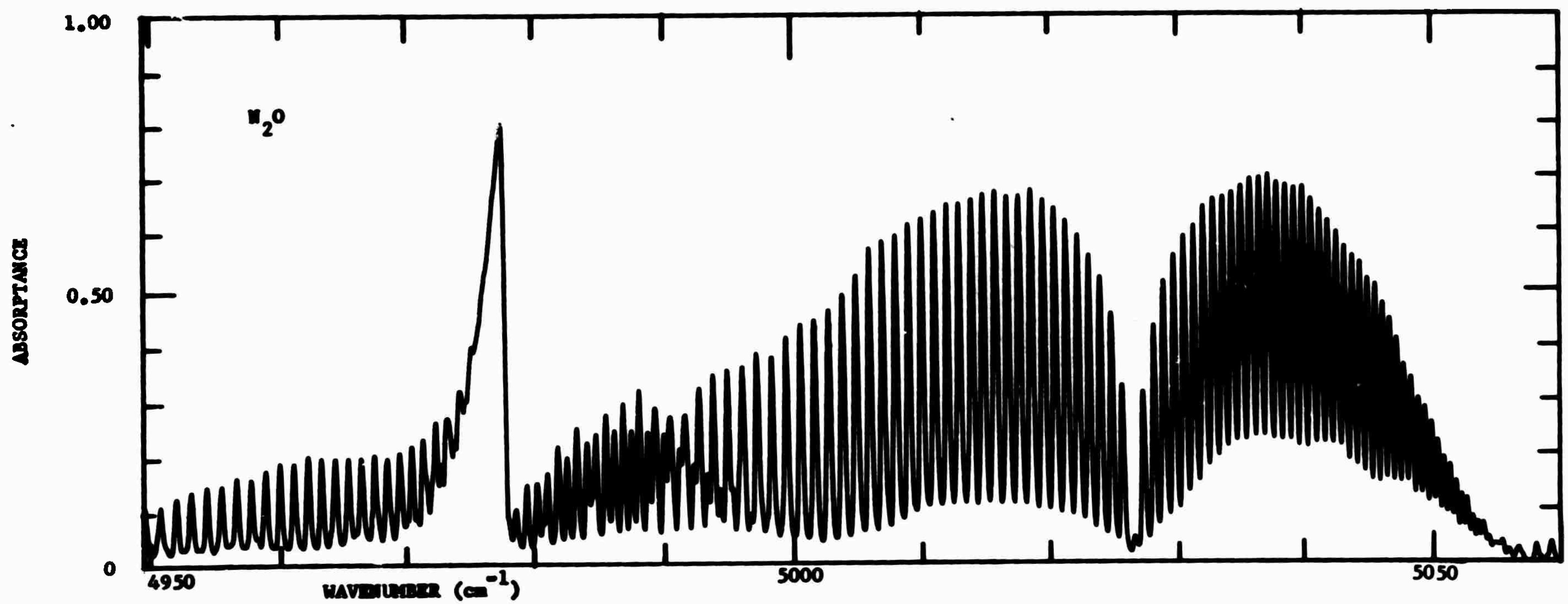

FIG. 9. Spectral curve of absorptance between 4950 and 5060 cm^{-1} for a pure N_2O sample. u = 163E20 molecules cm^{-2}; p = 0.132 atm; L = 3290 cm; θ = 196 K. Spectral slitwidth ≅ 0.37 cm^{-1}.

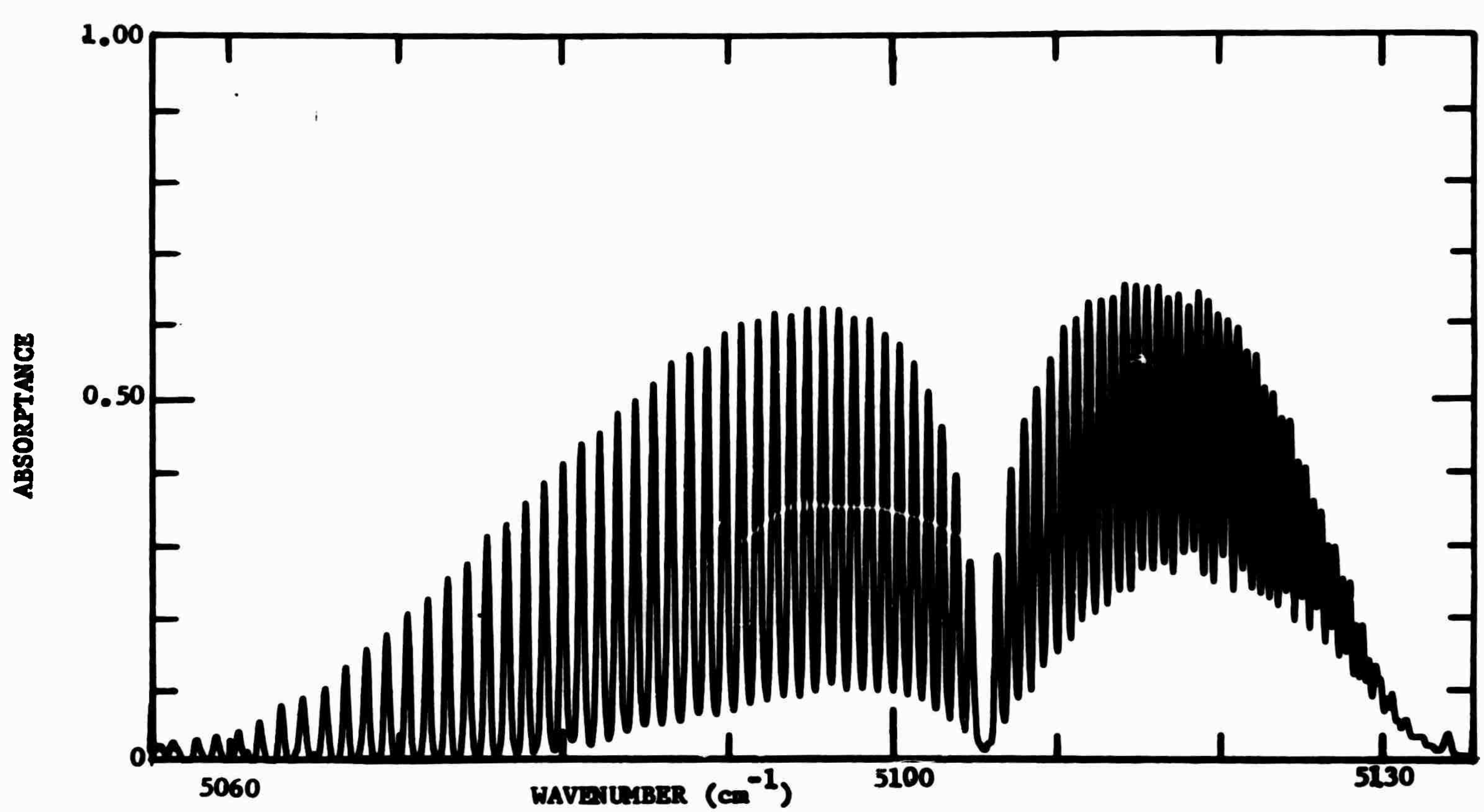

FIG. 10. Spectral curve of absorptance between 5055 and 5135 cm^{-1} for a pure N_2O sample. u = 163E20 molecules cm^{-2}; p = 0.132 atm; L = 3290 cm; θ = 196 K. Spectral slitwidth ℓ = 0.38 cm^{-1}.

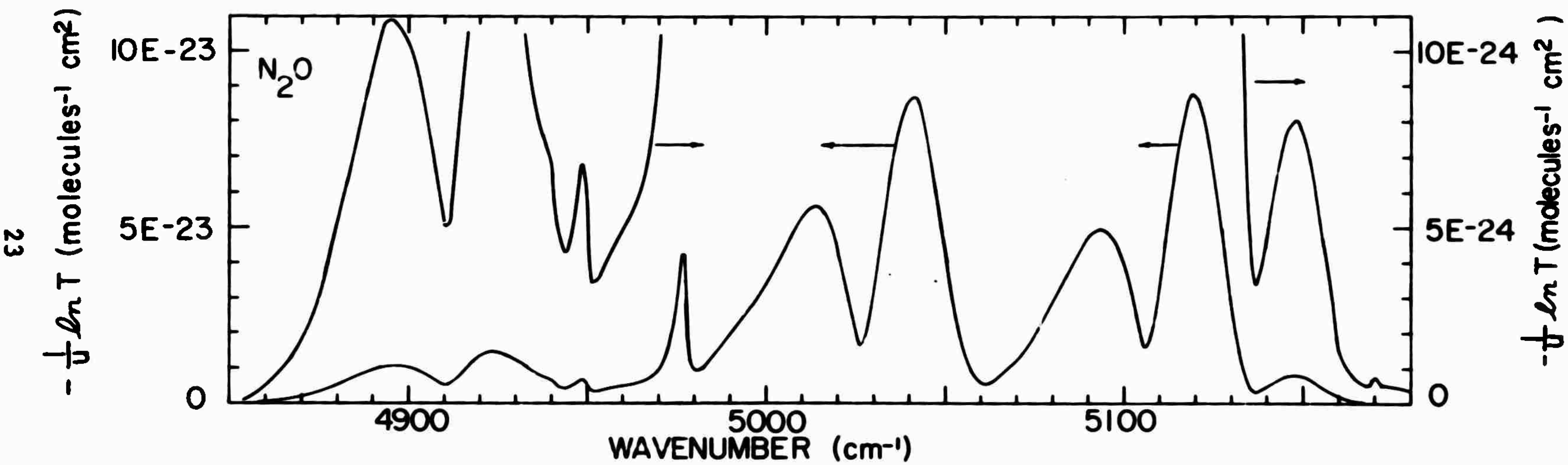

FIG. 11. Spectral curve of $(-1/u) \ln T$ between 4850 and 5180 cm^{-1} for an $N_2O + N_2$ sample at approximately 15 atm. $\theta \doteq 296$ K. Spectral slitwidth $\hat{=}$ 0.60 cm^{-1}. The arrows indicate the ordinate scale to be used.

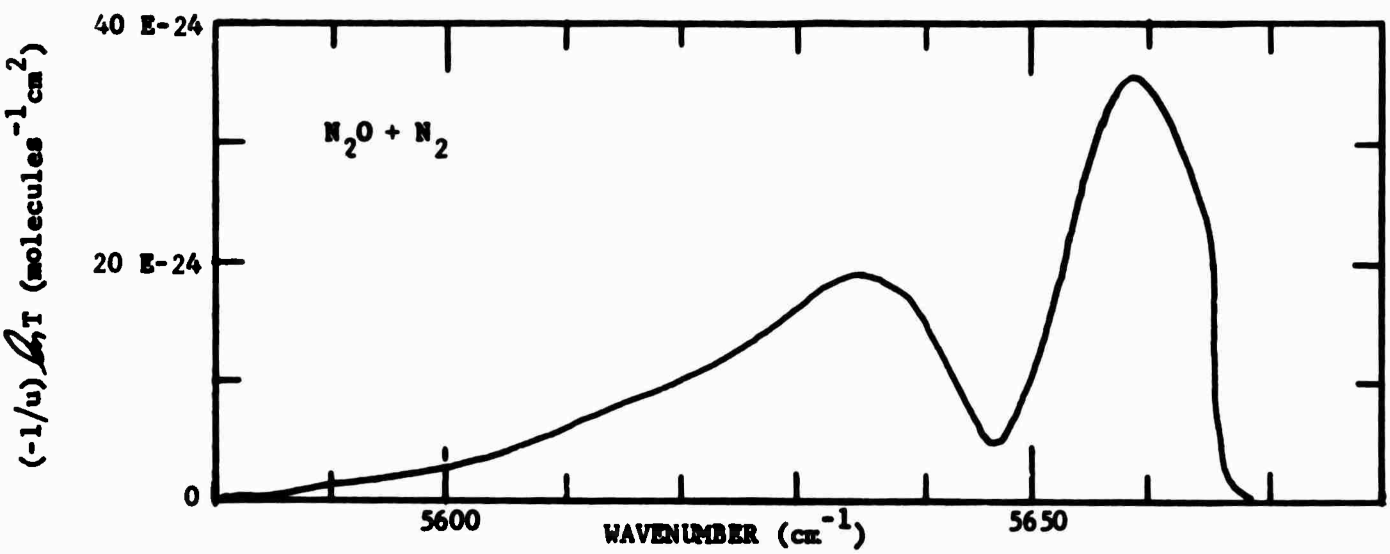

FIG. 12. Spectral curve of (-1/u) $\ln$ T between 5580 and 5680 cm^{-1} for an $N_2O + N_2$ sample at approximately 15 atm. Spectral slitwidth $\lessapprox$ 0.80 cm^{-1}. θ = 296 K.

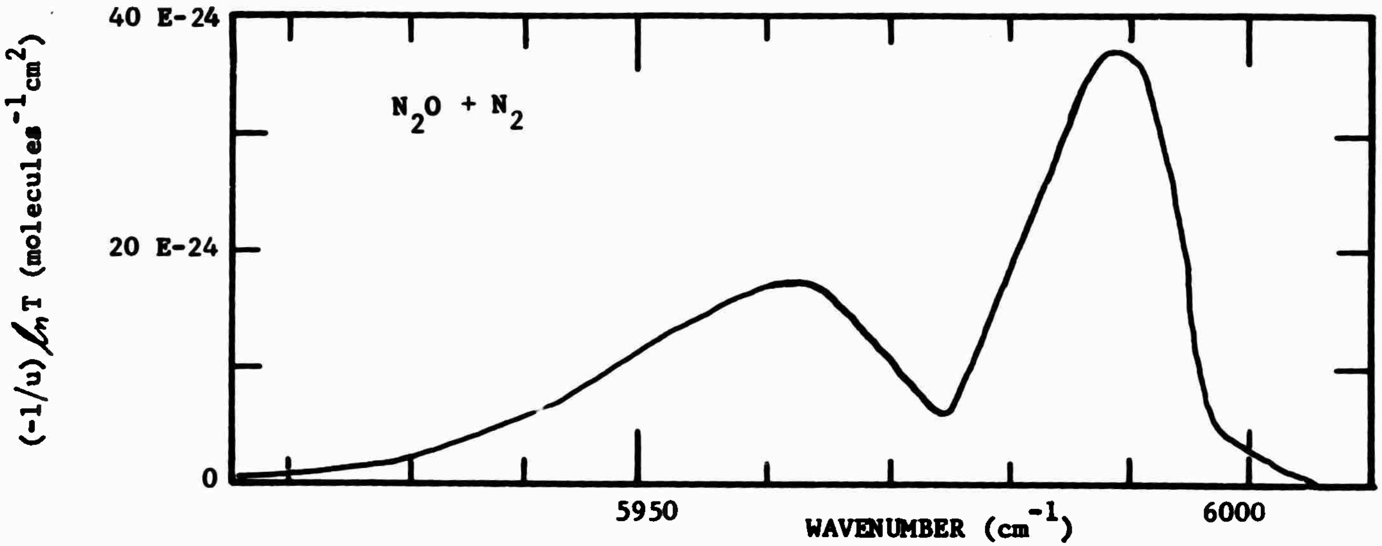

FIG. 13. Spectral curve of $(-1/u) \ln T$ between 5915 and 6010 cm^{-1} for an $N_2O + N_2$ sample at approximately 15 atm. Spectral slitwidth $\cong$ 0.92 cm^{-1}. θ = 296 K.

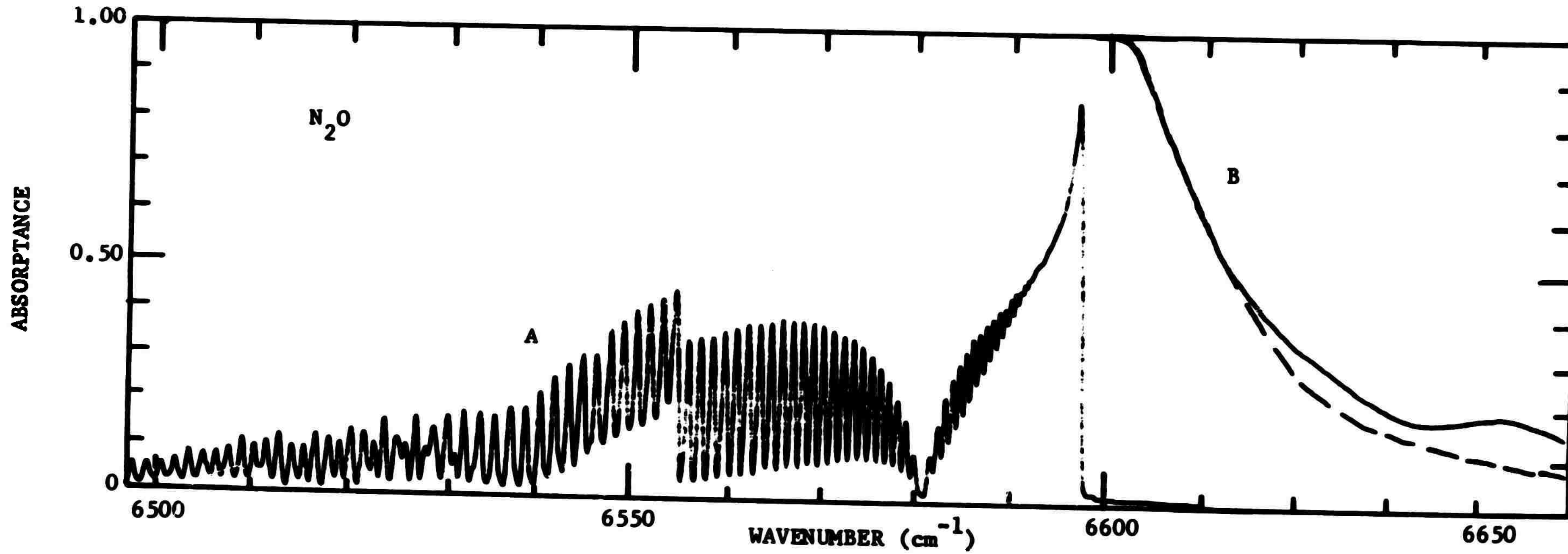

FIG. 14. Spectral curves of absorptance between 6500 and 6650 cm^{-1} for two samples of pure N_2O. The absorptance of Sample B is 1 between 6500 and 6600 cm^{-1}. The broken curve between 6610 and 6650 cm^{-1} represents the estimated absorptance by the wings of the lines of the 00^03 band. The difference between the broken line and the solid one is due to absorption by very weak lines centered between 6610 and 6650 cm^{-1}. Spectral slitwidth $\cong 0.7$ cm^{-1}.

Sample Number	u (molecules cm^{-2})	p (atm)	L (cm)	θ (Kelvin)
A	2.04 E 22	0.25	3290	296
B	1.36 E 24	14.6	3290	296

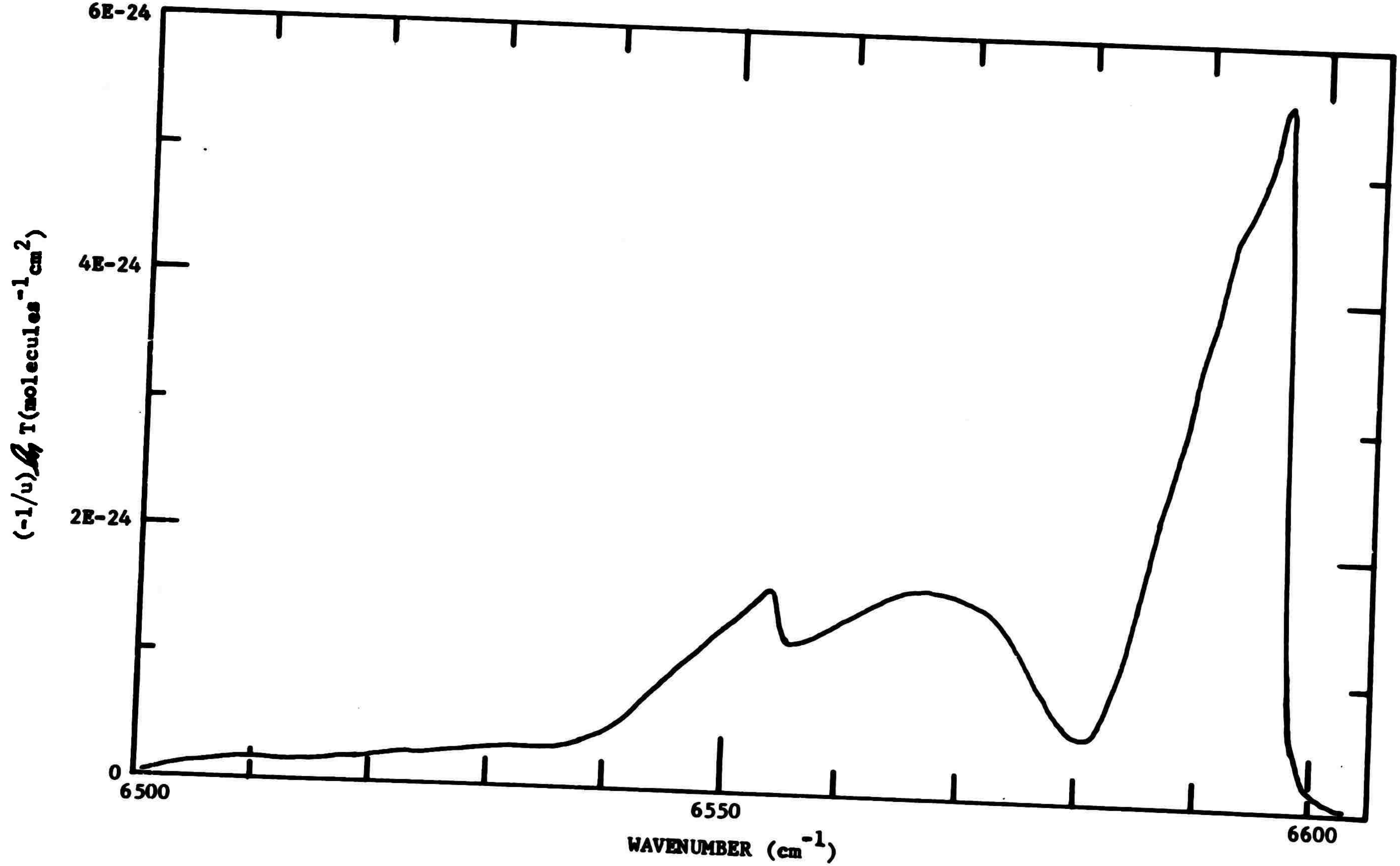

FIG. 15. Spectral curve of $(-1/u) \ln T$ between 6500 and 6605 cm^{-1} for an N_2O sample at approximately 15 atm. $\theta = 296$ K. Spectral slitwidth = 0.7 cm^{-1}.

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